β- and γ-counting for pre-detonation nuclear forensics on Eu-155

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Post-detonation nuclear forensics was performed at Los Alamos National Laboratory (LANL) on ^{155}Eu, a fission product on the wing of the fission product production curve whose yield is sensitive to fission fuel and neutron energy. With a half-life of 4.753 years, ^{155}Eu provides a longer-lived option for these measurements than other fission products with similar mass numbers. The Chemistry Division Group - Nuclear and Radiochemistry, at LANL routinely measures a suite of fission products from ^{235}U fissions in thermal neutron flux experiments known as thermal calibration exercises, using a mixture of gas proportional β-decay counting and γ-spectrometry on HPGe detectors. The fission products of interest are reported relative to a high-yield reference fission product from the same sample to create a running-average ratio specific to neutron energy and fuel type; Equation 1 below shows the ratio-of-ratios R-value measured fission products are reported in:

$$R_{^{i}X} = \frac{\left[A(^{i}X)/A(^{99}Mo)\right]_{unknown}}{\left[A(^{i}X)/A(^{99}Mo)\right]_{^{235}U_{n,th}}} \quad (1)$$

where A denotes activity, ^{i}X is the nuclide of interest, and $^{235}U_{n,th}$ denotes irradiations of ^{235}U with thermal neutrons [1]. Measurements of unknown fission spectra are ratioed to a running-average of thermal calibration results, and the resulting R-value can be referenced against a library of irradiation conditions. This work adapted the existing methodology to ^{155}Eu and preliminary results are shown here. These results are in preparation for publication.

Both β-counting and γ-spectrometry were assessed for their viability in measurements of ^{155}Eu. β-decay counts ranged from 0.1 to 20 counts per minute when detectable, owing to several factors: low specific activity, low fission product yield, potential trace contaminants and a low β-decay energy, all of which hampered β-counting. The method is extremely sensitive to background fluctuations, with multiple available samples not distinguishable above background, and thus β-counting was determined to be unreliable for ^{155}Eu without considerable extra R&D. A method using only γ-spectrometry was developed, which can use running-averages of count rates on the same detector or can be converted to activity for comparison across detectors.

After thermal calibration samples were analyzed to create a running-average value for the denominator in Eq. 1, preliminary results using only γ-spectrometry to characterize non-standard irradiations were compiled, as shown in Figs. 1 and 2. Twelve separate samples representing six unique irradiation conditions were analyzed, using ^{235}U, ^{238}U, and ^{239}Pu fuels with three different neutron fields: thermal, fission and 14 MeV fusion; additionally, one experiment induced fission with a proton beam on natural uranium. Fig. 1 shows the greater precision this method provides over calculating expected values based on published fission product yields from the JEFF-3.1.1 database, where literature on the irradiation conditions even exists [2]. Literature values shown are cumulative fission yields for different irradiation conditions reported as R-values for direct comparison. Due to large uncertainties on the fission yields of ^{155}Eu, the literature data show significantly worse precision than the measured values.

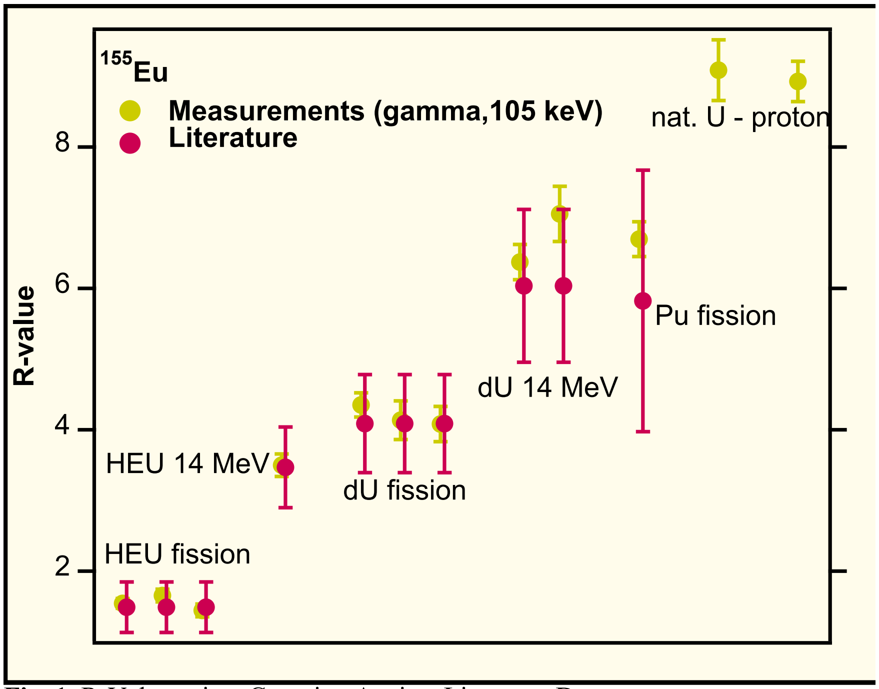

Fig. 1. R-Values via γ-Counting Against Literature Data.

Fig. 2 shows that the method produces values between those of ^{156}Eu and ^{153}Sm. Increasing fuel mass or neutron energy tends to systematically change R-values in correlation to the mass number of the fission product; this would indicate that R-values for ^{155}Eu should be found between ^{153}Sm and ^{156}Eu, and

Nuclide	Analysis Method
^{156}Eu	Beta Counting
^{155}Eu	Gamma
^{153}Sm	Beta Counting

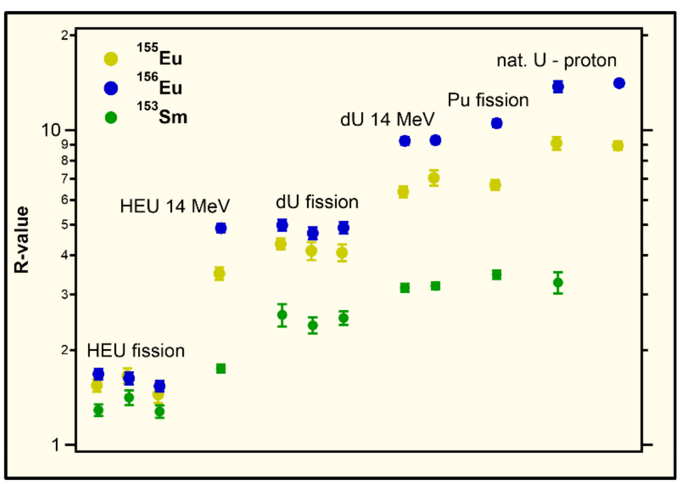

Fig. 2. Results of ^{155}Eu Analyses on Non-Standard Irradiations.

Fig. 2 shows that this relationship holds across several unique fission fuels and neutron energies. ^{153}Sm and ^{156}Eu are short-lived nuclides previously analyzed for these irradiations.

This study has been cleared for release by LANL (LA-UR-23-28541). As a result of this work, ^{155}Eu measured by γ-spectrometry can be utilized in future irradiation campaigns.

[1] M.J. Jackson *et al.*, J. Radioanal. Nucl. Chem. **318**, 107 (2018). doi:10.1007/s10967-018-6048-1
[2] The JEFF-3.1/-3.1.1 radioactive decay data and fission yields sub-libraries, JEFF report 20.